

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Takuo SUGIOKA et al.

Serial No.: 10/516,695 Art Unit: 1796

Filed : January 6, 2005 Examiner: ANTHONY, JOSEPH DAVID

Title : FIRE RETARDANT RESIN COMPOSITION, METHOD OF ITS
PRODUCTION, SHAPED ARTICLES COMPRISING THE SAME, AND SILICA

DECLARATION UNDER RULE 1.132

Honorable Commissioner of Patents and Trademarks,
Washington, D.C. 20231

Sir:

I, Takuo Sugioka, a citizen of Japan and having
postal mailing address of Koyoen-megamiyama 31-74
Nishinomiya, Hyogo, JAPAN, declare and say that:

March 1995, I was graduated from Osaka University
and received Master degree of Material Science and
Chemistry;

From April 1995, up till the present, I have been
employed by Nippon Shokubai Co., Ltd., and engaged in the
works of Electronics-Related material Sceience Research;

I am one of the inventors of the above-identified
application and am familiar with the subject matter
thereof;

I respectfully submit herewith my exact report
thereon;

Experiment for showing that the fire retardant resin composition of the present invention is excellent in fire retardancy as compared to a resin composition comprising a phenolic resin consisting of a structure where aromatic units having phenolic hydroxyl groups are connected to one another through a methylene group containing one carbon atom

(1) Preparation of a phenolic resin (Polyphenol Z)

Into a 2L four-necked flask equipped with a gas inlet, a Dean-Stark trap, and a stirring bar, phenol novolak resin (trade name "TD-2131", product of DAINIPPON INK AND CHEMICALS, INC.) 620 g and methanol 334 g were charged. Then, the mixture was stirred at 40°C to provide a completely homogeneous solution. Then, into the reaction liquid in the four-necked flask, two PTFE tubes were inserted. While the temperature was kept at 40°C, tetramethoxysilane 846 g and water 200 g were separately charged through the different tubes for 4 hours using a roller pump. After completion of the addition, the solution was kept at 60°C for 4 hours. Under nitrogen flow, the temperature was increased again, and residual water and methanol which began to be distilled off at near 80°C were trapped into the trap while stirring was performed until the temperature was increased to 160°C. Then, a volatile content was removed and the obtained substance was cooled. As a result, a milky solid polyphenol Z was obtained. With the milky solid polyphenol Z, the yield was 950 g; the thermal softening temperature was 97°C; the hydroxyl value was 158 g/mol; and the content of the inorganic compound was 35.1%.

(2) Preparation of molded product (Additional Comparative Example)

A cured article was produced in the same manner as in Example 7 on page 55, line 33 to page 56, line 18 in the specification, except that the above-mentioned polyphenol Z instead of the polyphenol composition A. Then, this cured article was evaluated for mechanical and thermal properties and fire retardancy. Table A shows the results.

Table A also includes results in Examples 7 to 12 and Comparative Examples 3 and 4 on page 57 in the specification, for reference.

(3) Conclusion

The results in Table A clearly show that the resin composition in Additional Comparative Example, which was obtained using a phenolic resin consisting of a structure where aromatic units having phenolic hydroxyl groups are connected to one another through a methylene group containing one carbon atom, doesn't provide a cured article having a fire retardancy needed for practical use.

Accordingly, it is proven that the resin composition including the polyphenol compound having a specific structure, disclosed in the present application, has much more excellent fire retardancy than that of the resin composition including the phenolic resin.

Table A

	Example	Comparative Example				Additional Comparative Example				
		7	8	9	10	11	12	3	4	
EPN-1180	48.3	46.9	43.4	45.1	49.3	48.2	54.2	51.3	53.3	
Polyphenol composition A	51.7	—	—	—	—	—	—	—	—	
Polyphenol composition B	—	53.1	—	—	—	—	—	—	—	
Polyphenol composition C	—	—	56.6	—	—	—	—	—	—	
Polyphenol composition D	—	—	—	54.9	—	—	—	—	—	
Polyphenol composition E	—	—	—	—	50.7	—	—	—	—	
Polyphenol composition F	—	—	—	—	—	51.8	—	—	—	
Polyphenol composition G	—	—	—	—	—	—	45.8	—	—	
Polyphenol composition H	—	—	—	—	—	—	—	48.7	—	
Polyphenol Z	—	—	—	—	—	—	—	—	46.7	
2E4MZ	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	
Flexural strength /MPa	103	125	118	130	110	100	79	109	108	
Flexural elastic modulus/GPa	4.58	4.58	4.20	4.79	4.80	4.28	3.62	4.06	3.98	
Fracture toughness/MPa·m ^{1/2}	0.77	0.75	0.86	0.67	0.75	0.81	0.75	0.71	0.78	
Tg(TMA)/°C	130	129	125	133	143	130	130	129	142	
UL-94 fire retardancy	Average flame-out time/sec	3.8	1.0	2.2	1.5	0.8	1.2	Combusted	58	Combusted
Evaluation	V-0	V-0	V-0	V-0	V-0	V-0	—	V-1	—	

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 14th the day of July, 2008

Takuo Sugioka

Takuo Sugioka

14th of July, 2008
Takuo Sugioka